



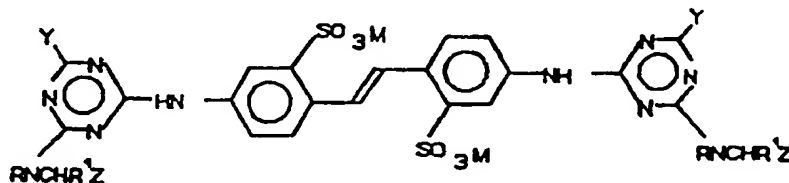
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<p>(21) International Application Number: PCT/GB95/01428</p> <p>(22) International Filing Date: 19 June 1995 (19.06.95)</p> <p>(30) Priority Data: 9412756.0 24 June 1994 (24.06.94) GB</p> <p>(71) Applicant (for all designated States except US): HICKSON INTERNATIONAL PLC [GB/GB]; Chancellor Court, 21 The Calls, Leeds LS7 2HG (GB).</p> <p>(72) Inventor; and</p> <p>(75) Inventor/Applicant (for US only): THOMPSON, John, Dennis [GB/GB]; Hickson Fine Chemicals, Wheldon Road, Castleford, West Yorkshire WF10 3JT (GB).</p> <p>(74) Agent: SKAILES, Humphrey, John; Frank B. Dehn & Co., Imperial House, 15-19 Kingsway, London WC2B 6UZ (GB).</p>	<p>(81) Designated States: BR, CZ, KR, PL, RU, US, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).</p> <p>Published</p> <p><i>With international search report.</i> <i>Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i></p>	

(54) Title: S-TRIAZINYLAMINOSTILBENE DERIVATIVES AND THEIR USE AS OPTICAL WHITENING AGENTS**(57) Abstract**

S-Triazinylaminostilbene derivatives of formula (1) are described in which Y is halogen, alkoxy or -NR²R³ (where R² and R³ are the same or different and are H, alkyl, hydroxyalkyl or aryl or together with the nitrogen atom form a substituted or unsubstituted cyclic amino group having 5-7 ring members), R is H, aryl, which is

unsubstituted or substituted (e.g. by one or more electron releasing groups such as C₁₋₄alkyl and/or C₁₋₄alkoxy), R¹ is H or alkyl, Z is an electron-withdrawing group (e.g. CN, -SO_xM), x is 2 or 3 and M is H, a metal cation or -N⁺R⁴R⁵R⁶R⁷ (where R⁴-R⁷ are the same or different and are H, alkyl, aralkyl or hydroxyalkyl). The compounds are useful as optical whitening agents for paper or textiles.



(1)

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5 The present invention relates to novel \bar{s} -triazinylaminostilbene derivatives, processes and intermediates for their preparation and their use as optical whitening agents (OWAs) for whitening natural, synthetic and semi-synthetic polymers.

10 The area of use covered by \bar{s} -triazinylaminostilbene OWAs is very broad, but of particular interest is the whitening of paper and textile substrates by the class of triazinylaminostilbene derivatives known as the "hexasulphonated" compounds resulting from the use of aniline-2,5-disulphonic acid as one of the substituents. The substantivity (i.e. the natural affinity of the substance for the fibre) of this class of OWA to cellulose for example, is generally a function of the number of
15 solubilising groups (particularly sulphonic acids) present in the molecule: thus, the disulphonic acid derivatives, such as 4,4'-bis(6"-phenylamino-4"-morpholino-1",3",5"-triazin-2"-yl)aminostilbene-2,2'-disulphonic acid, are very substantive, the tetrasulphonic acid derivatives, such as 4,4'-bis[6"- (4'''-sulphophenylamino)- 4"-morpholino-1",3",5"-triazin-2"-yl]aminostilbene-2,2'-disulphonic acid, are
20 moderately substantive, and the hexasulphonic acid derivatives, such as 4,4'-bis[6"- (2''',5'''-disulphophenylamino)-4"-morpholino-1",3",5"-triazin-2"-yl]aminostilbene-2,2'-disulphonic acid, are hardly substantive.

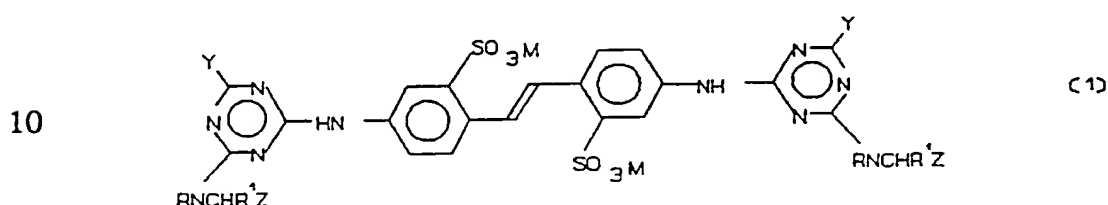
We have now found a new group of \bar{s} -triazinylaminostilbenes which contain a
25 solubilising group (such as sulphonic acid) attached to a methylamino substituent on the triazine ring. This contrasts to the tetra- and hexasulphonic acid derivatives referred to above in which the sulphonic acid substituents are attached to phenylamino groups. Surprisingly, these compounds are stable to acid hydrolysis and have different properties compared to the known tetrasulphonic acid derivatives. In
30 particular we have found that these tetrasulphonic acid derivatives have the substantivity properties of the hexasulphonic acid derivatives of the prior art OWAs. These tetrasulphonic acids are thus especially useful for application to paper either alone, or in combination with other optical whitening agents, dyestuffs, pigments or

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other additives by size press or other surface application methods, and to textiles by padding, either alone or in combination with other typical textile finishing additives e.g. acid resins, crease- and water-proofing agents, or specialist finishes, or during laundering processes as part of detergent or fabric conditioner compositions.

5

The novel *s*-triazinylaminostilbene derivatives according to the invention are compounds of formula (1)



in which:

15 Y is halogen (e.g. fluoro, chloro or bromo), alkoxy (e.g. C₁₋₄ alkoxy) or -NR²R³ [where R² and R³ are the same or different and are H, alkyl (e.g. C₁₋₄alkyl), hydroxyalkyl (e.g. C₁₋₄hydroxyalkyl), or aryl (e.g. phenyl, optionally substituted), or together with the nitrogen atom form a substituted or unsubstituted cyclic amino group having 5-7 ring members, e.g. morpholino or piperidino],

20 R is H, aryl (e.g. phenyl), which is unsubstituted or substituted (e.g. by one or more electron releasing groups such as C₁₋₄alkyl and/or C₁₋₄alkoxy),

R¹ is H or alkyl (e.g. C₁₋₄alkyl, optionally substituted),

25 Z is an electron-withdrawing group (e.g. CN, -SO_xM),

x is 2 or 3 and

30 M is H, a metal cation (e.g. alkali metal, such as Na, K or Li) or -N⁺R⁴R⁵R⁶R⁷ (where R⁴-R⁷ are the same or different and are H, alkyl (e.g. C₁₋₄alkyl), aralkyl (e.g. phenyl(C₁₋₆)-alkyl) or hydroxyalkyl (e.g. C₁₋₄ hydroxyalkyl)).

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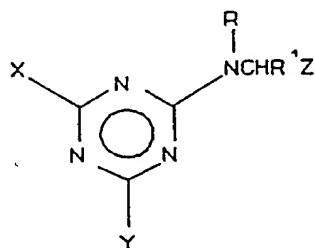
Y is preferably bis(2-hydroxyethyl)amino or morpholino. R is preferably phenyl or substituted phenyl, e.g. *o*-, *m*- or *p*-tolyl, anisyl, xylyl or cresyl. R¹ is preferably H or -CH₃ and Z is preferably -SO₃M (particularly where M is an alkali metal cation such as Na) or CN.

5

A compound of particular importance is the compound in which Y is bis(2-hydroxyethyl)amino, R is phenyl, R¹ is H and Z is -SO₃Na.

10

The compounds of formula (1) can be prepared by reacting a compound of formula (2)



(2)

15

(where X is halogen (e.g. fluoro, chloro or bromo), Y is halogen (e.g. fluoro, chloro or bromo) or alkoxy (e.g. C₁₋₆alkoxy) and N(R)CHR¹Z is as defined above) with 4,4'-diaminostilbene-2,2'-disulphonic acid.

20

To ensure complete reaction, at least 2 moles of the compound (2) should be added per mole of the disulphonic acid. The reaction can be carried out at any convenient temperature (e.g. 20-60°C or to reflux) in a suitable solvent (e.g. water or aqueous acetone), preferably in the presence of an acid binding agent such as sodium bicarbonate or carbonate.

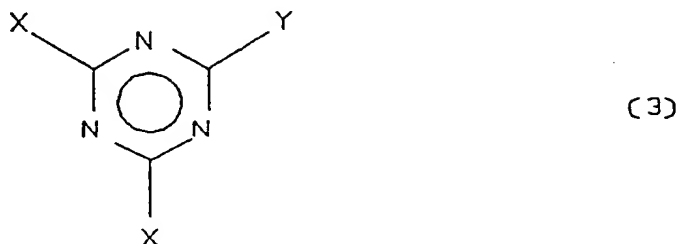
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When a compound in which Y is -NR²R³ is required, a compound of formula (1) in which Y is halogen can be reacted with an amine HNR²R³, in generally the same manner as for the preparations of other compounds of formula (1).

30

The compounds of formula (2), which also form part of the invention, may be

prepared by reacting a cyanuric halide of formula (3)



(where X and Y are as defined for formula (2) above) with an amine of formula (4), RNHCHR^1Z . This reaction may be performed as described above for the preparation of the compounds (1), preferably at 0-25°C.

10

The amines (4) may be prepared by reacting an amine RNH_2 with an alcohol HOCHR^1Z , for example in a neutral aqueous or aqueous alcoholic medium at 20-60°C.

15

Alternatively, the compounds of formula (4), ($\text{Z}=\text{CN}$) may be prepared from the compound of formula (4) ($\text{Z}=\text{SO}_3\text{Na}$) by treatment with a soluble cyanide (e.g. NaCN , KCN).

20 The main applications of the compounds of formula (1), particularly the preferred compounds, lie in the optical whitening of paper and textiles under conditions normally used for the "hexasulphonic acid derivatives" referred to above. However, they may also be used to treat any other cellulosic or other natural, synthetic or semi-synthetic materials to which optical brightening is relevant.

25 The following Examples illustrate the invention.

EXAMPLE 1

30 Tetra-sodium 4,4'-bis[6"-N-phenyl-N-sulphonatomethylamino-4"-bis(2'''-hydroxyethyl)amino-1",3",5"-triazin-2"-yl]aminostilbene-2,2'-disulphonate

- 5 -

To a solution of sodium N-phenyl-N-(2,4-dichloro-1,3,5-triazin-6-yl)aminomethane sulphonate (37.5g, 0.105g. mol.) in aqueous acetone (375ml) at 20°C containing sodium bicarbonate (8.8g, 0.105 g.mol.) is added a solution of sodium 4,4'-diaminostilbene-2,2'-disulphonate (20.7g, 0.05 g.mol.) in water (140ml) at 20°C.

5 The mixture is then heated up to 50°C and maintained at that temperature until the reaction is complete (ca. 1 hour). Diethanolamine (12.1g, 0.115 g.mol.) and sodium bicarbonate (8.8g, 0.105 g.mol.) are then added and the solution is heated to 100°C whilst distilling off the acetone and maintained at that temperature for 2 hours. The resulting solution may be used as is, or may be desalinated and concentrated by

10 ultrafiltration to give the title compound as a concentrated solution.

The starting material is prepared as follows:

Sodium N-phenyl-N-(2,4-dichloro-1,3,5-triazin-6-yl) aminomethane sulphonate

15 A solution of cyanuric chloride (19.4g, 0.105.mol.) in acetone (200ml) containing sodium bicarbonate (8.8g, 0.105g.mol.) is cooled to -10°C, and an ice-cold solution of sodium ω -anilinomethane sulphonate (23.0g, 0.110g.mol.) in water (175ml) is added as rapidly as possible and the mixture is allowed to warm up to room

20 temperature. The reaction may be followed by means of HPLC using detection at 265nm to enable visualisation of all the starting products and final products, (including any by-products) to be observed. The resulting solution of the title compound may then be used for further condensations with nucleophiles, e.g. 4,4'-diaminostilbene-2,2'-disulphonic acid and other amines as required.

25 The solution of sodium ω -anilinomethane sulphonate is prepared as follows. Aniline (10.2g, 0.110 g.mol) is added to a stirred solution of the sodium salt of the formaldehyde bisulphite addition complex (15.4g, 0.115 g.mol) in water (100ml) at 25-30°C. Formation of the sodium phenylaminomethane sulphonate is complete

30 within 90 minutes (less at higher temperatures) and the solution may then be diluted and cooled and used as described above.

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EXAMPLE 2

Tetra-sodium 4,4'-bis[6"-N-phenyl-N-sulphonatomethylamino-4"-morpholino-1",3",5"-triazin-2"-yl]aminostilbene-2,2'-disulphonate

5

The above compound is prepared by the method of Example 1 except that the diethanolamine is replaced with morpholine (10.0g, 0.115g. mol.) to give the corresponding morpholine derivative. The product may be isolated by salting followed by filtration.

10

EXAMPLES 3-6

15

The aniline used in the preparation of the starting material in Examples 1 and 2 may be replaced by p-toluidine (11.8g, 0.110g.mol.) or p-anisidine (13.5g, 0.110g.mol.) to give the corresponding derivatives.

The following is a further example of a compound of formula (2).

EXAMPLE 7

20

N-Phenyl-N-(2,4-dichloro-1,3,5-triazin-6-yl) aminoacetonitrile

25

Cyanuric chloride (18.5g) is added all at once to a stirred, ice-cold slurry of phenylaminoacetonitrile (13.9g) in water (150ml) containing sodium bicarbonate (8.4g) and the mixture is allowed to warm up to room temperature. The resulting slurry of the title compound may then be used for further condensations with nucleophiles, e.g. 4,4'-diaminostilbene-2,2'-disulphonic acid and other amines as required.

30

The slurry of phenylaminoacetonitrile is prepared as follows. Aniline (9.8g) is added to a stirred solution of glycolonitrile (55% aqueous solution, 11.5g) in water (100ml)

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at 20°C, warmed up to 60°C and maintained at that temperature until the reaction is complete (ca. 1 hour). The resulting oily dispersion may then be cooled and diluted and used as described above.

- 5 The following Examples illustrate the use of the compounds of Examples 1 and 2.

Examples A - Size Press Application

Blank handsheets (comprised of 27.2% bleached kraft softwood, 52.8% bleached
10 kraft hardwood and 20% calcium carbonate filler internally sized with alkyl ketene
dimer) are passed through 9% clear starch solution at pH 6 containing 30, 40 and 50
g/l of OWA (optical strength ($E_{1cm}^{1\%}$)62) and then fed through a size press to squeeze
out the excess liquor. The OWAs used are (i) the compound of Example 1 and (ii)
Photine^(R) SLC Standard (a proprietary brand of "hexa-sulpho" OWA). The sheets
15 are dried (4 minutes at 105°C), conditioned (1 hour at room temperature and 70%
R.H.) and then the fluorescence is measured on a Harrison Meter and the brightness
on an ICS Q MM 2000 meter, with and without UY filter. To check on the shade
and overall appearance, a visual assessment of the two sets of patterns is also made.
Typical readings of brightness lift (e-f) obtained are:-

20

Concentration (g/l of $E_{1cm}^{1\%}$ 62)	Product from Example 1	Photine ^(R) SLC
30	13.1	15.9
40	16.0	16.9
50	17.3	16.8

25

Visual Assessment

30

Equivalent to the Photine^(R) SLC patterns.

Example B - Padding Application

Patterns of 80 x 80 bleached cotton fabric are passed through solutions of OWAs containing 5, 10, 15 and 20 g/l of OWA of optical strength ($E_{1cm}^{1\%}$) adjusted to 65, and then fed through a padding mangle at constant pressure to give 100% wt/wt pick-up of liquor. Where a resin finish is employed, the padding solutions contain (in addition to the OWA) 100 g/l Fixapret^(R) CPN and 10 g/l magnesium chloride. The OWAs used are (i) that of Example 2 and (ii) a "hexa-sulpho" OWA based on aniline-2,5-disulphonic acid and morpholine. The patterns are dried for 5 minutes at 140°C and conditioned for 1 hour at room temperature and 70% R.H. The fluorescence is then measured using a Harrison fluorescence meter and the ISO whiteness by means of a ICS Q MM 2000 meter. Typical readings obtained are:-

(a) Harrison Meter

Concentration (g/l. of $E_{1cm}^{1\%}$ 65)	Water		Resin	
	Product from Example 2	Hexa-sulpho OWA	Product from Example 2	Hexa-sulpho OWA
5	309	316	317	335
10	338	347	362	366
15	365	373	382	383
20	369	396	391	386

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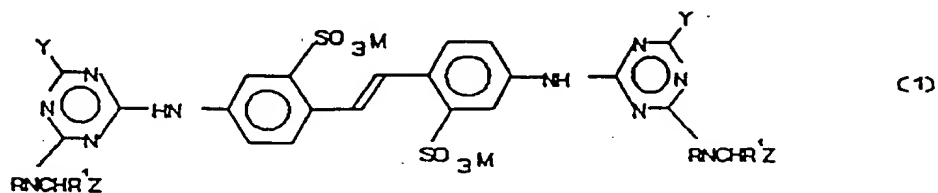
(b) ISO Brightness

Concentration (g/l of $E_{1cm}^{1\% 65}$)	Water		Resin	
	Product from Example 2	Hexa-sulpho OWA	Product from Example 2	Hexa-sulpho OWA
5	111.9	113.1	111.5	112.1
10	113.8	114.7	114.9	115.0
15	115.9	117.5	115.8	116.8
20	116.4	119.3	115.5	115.9

- 10 These results show that the compound of Example 2 is comparable in performance to the hexa-sulpho compound.

CLAIMS:

1. s-Triazinylaminostilbene derivatives of formula (1)



in which:

Y is halogen, C₁₋₄ alkoxy or -NR²R³ (where R² and R³ are the same or different and are H, C₁₋₄alkyl, C₁₋₄hydroxyalkyl or phenyl (optionally substituted), or together with the nitrogen atom form a substituted or unsubstituted cyclic amino group having 5-7 ring members),

R is H, or phenyl, which is unsubstituted or substituted by one or more electron releasing groups such as C₁₋₄alkyl and/or C₁₋₄alkoxy,

R¹ is H or C₁₋₄alkyl, optionally substituted,

Z is an electron-withdrawing group such as CN or -SO_xM,

x is 2 or 3 and

M is H, a metal cation or -N⁺R⁴R⁵R⁶R⁷ (where R⁴-R⁷ are the same or different and are H, C₁₋₄alkyl, phenyl(C₁₋₆)-alkyl or C₁₋₄ hydroxyalkyl).

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2. Compounds as claimed in claim 1 in which:

Y is bis(2-hydroxyethyl)amino or morpholino,

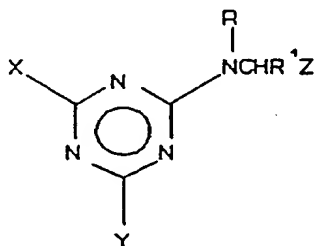
R is phenyl, *o*-, *m*- or *p*-tolyl, anisyl, xylyl or cresyl,

R¹ is H or -CH₃ and

Z is -SO₃M (where M is an alkali metal cation such as Na) or CN.

3. A compound as claimed in claim 1 in which Y is bis(2-hydroxyethyl)amino, R is phenyl, R¹ is H, Z is -SO₃Na and M is Na.

4. A process for the preparation of a compound as claimed in claim 1 which comprises reacting a compound of formula (2)



(2)

(where X is halogen, Y is halogen or C₁₋₆alkoxy and N(R)CHR¹Z is as defined in claim 1) with 4,4'-diaminostilbene-2,2'-disulphonic acid.

5. A composition for the whitening of paper or textiles which contains a compound as claimed in claim 1 as an optical whitening agent.

6. A composition as claimed in claim 5 which includes an aqueous medium.

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7. The use of a compound as claimed in any of claims 1 to 3 for the whitening of natural, synthetic or semi-synthetic polymeric materials.

8. The use of a compound as claimed in any of claims 1 to 3 for application to paper, for example by surface application from an aqueous medium which can include starch, carboxymethylcellulose, polyvinyl alcohol or other natural or synthetic polymeric binders.

9. The use of a compound as claimed in any of claims 1 to 3 for application to textiles, for example by padding, either alone or in combination with other textile finishing additives.

10 The use of a compound as claimed in any of claims 1 to 3 as an optical brightening additive in a laundry detergent, for example a liquid detergent, or in a fabric conditioner composition.

INTERNATIONAL SEARCH REPORT

 Interna: I Application No
 PCT/GB 95/01428

A. CLASSIFICATION OF SUBJECT MATTER

 IPC 6 C07D251/68 C08K5/42 D06L3/12 D21H21/30 D21H19/36
 C11D3/42 C11D9/44 //C07D251/50, C07D251/52

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C07D C08K D06L D21H C11D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	DE-A-21 24 079 (IMPERIAL CHEMICAL INDUSTRIES LTD.) 2 December 1971 see claims 1,2,4,7,8; example 8 ---	1,4-10
X	CHEMICAL ABSTRACTS, vol. 81, no. 8, 26 August 1974 Columbus, Ohio, US; abstract no. 38963b, MATSUO M. ET AL. 'Stilbenedisulfonic acid fluorescent whiteners' page 91; column 2; see abstract & JP-A-49 010 216 (SUMITOMO CHEMICAL CO., LTD.) 29 January 1974 see compounds IX, X and VII see page 97 --- -/--	1,4-10

☒ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

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Date of the actual completion of the international search

13 October 1995

Date of mailing of the international search report

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INTERNATIONAL SEARCH REPORT

Internal Application No

PCT/GB 95/01428

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	CHEMICAL ABSTRACTS, vol. 68, no. 20, 13 May 1968 Columbus, Ohio, US; abstract no. 88211g, OKUBO I. ET AL. 'Stilbene fluorescent brightening agents' page 8524; column 2; see abstract & JP-A-42 021 014 (MITSUI CHEMICAL INDUSTRY CO., LTD.) 18 October 1967 ---	1-10
Y	CHEMICAL ABSTRACTS, vol. 69, no. 7, 12 August 1968 Columbus, Ohio, US; abstract no. 27392r, TEMKINA V.Y. ET AL. 'New fluorescent complexons in the bis(triazinyl-amino)stilbene series' page 2555; column 2; see abstract & TR. VSES. NAUCH.-ISSLED. INST. KHIM. REAKTIVOV OSOBO CHIST. KHIM. VESHCHESTV, no. 30, 1967 pages 131-144, ---	1-10
Y	CHEMICAL ABSTRACTS, vol. 80, no. 18, 6 May 1974 Columbus, Ohio, US; abstract no. 97352d, PIRKL J. 'Fluorescent whiteners' page 77; column 2; see abstract & CS-A-149 743 (PIRKL J.) 15 August 1973 ---	1-10
Y	CHEMICAL ABSTRACTS, vol. 81, no. 22, 2 December 1974 Columbus, Ohio, US; abstract no. 137605t, OHTAKI E. ET AL. 'Fluorescent whiteners' page 76; column 2; see abstract & JP-A-49 032 923 (MITSUI TOATSU CHEMICAL, INC.) 26 March 1974 see page 114; example NO.4 -----	1-10

Information on patent family members

PCT/GB 95/01428

Form PCT/ISA/210 (patent family annex) (July 1992)